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Measurements of Waste Tank Passive Ventilation Rates Using Tracer Gases

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September 1997

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

This report presents the results of ventilation rate studies of eight passively ventilated high-level radioactive waste tanks using tracer gases. Headspace ventilation rates were determined for Tanks A-101, AX-102, AX-103, BY-105, C-107, S-102, U-103, and U-105 using sulfur hexafluoride (SF_6) and/or helium (He) as tracer gases. Passive ventilation rates are needed for the resolution of several key safety issues. These safety issues are associated with the rates of flammable gas production and ventilation, the rates at which organic salt-nitrate salt mixtures dry out, and the estimation of organic solvent waste surface areas.

This tracer gas study involves injecting a tracer gas into the tank headspace and measuring its concentration at different times to establish the rate at which the tracer is removed by ventilation. Tracer gas injection and sample collection were performed by SGN Eurisys Service Corporation and/or Lockheed Martin Hanford Corporation, Characterization Project Operations. Headspace samples were analyzed for He and SF_6 by Pacific Northwest National Laboratory (PNNL).

The tracer gas method was first demonstrated on Tank S-102. Tests were conducted on Tank S-102 to verify that the tracer gas was uniformly distributed throughout the tank headspace before baseline samples were collected, and that mixing was sufficiently vigorous to maintain an approximately uniform distribution of tracer gas in the headspace during the course of the study. Headspace samples, collected from a location about 4 m away from the injection point and 15, 30, and 60 minutes after the injection of He and SF_6 , indicated that both tracer gases were rapidly mixed. The samples were found to have the same concentration of tracer gases after 1 hour as after 24 hours, suggesting that mixing of the tracer gas was essentially complete within 1 hour. Given this evidence for vigorous mixing, inhomogeneities produced by the influx of fresh air during normal ventilation would be expected to be restricted to a small region near the influx.

A comparison of He and SF_6 as tracer gases in the waste tanks was performed on five of the eight tanks. In two of the five tanks the concentration of SF_6 was found to decrease significantly faster than the concentration of He. The available evidence suggests that radiolytic decomposition of SF_6 occurred. A literature survey also revealed some indirect evidence suggesting that radiolysis effects associated with electron capture near the waste surface are credible, but the accelerated loss of SF_6 is not fully understood at this time. Because the stability of SF_6 in the waste tank headspaces was determined to be uncertain, SF_6 was not used in subsequent tank studies. However, in the three tanks where He and SF_6 concentrations decreased at essentially the same rate, the SF_6 results are considered to be valid, even for time periods where there are no He data.

Headspace ventilation rates were calculated from estimated headspace volumes and the decrease in tracer gas concentration over time. The calculated ventilation rate is consequently an average rate for the time period between sampling events. Periods between sampling events ranged from 4 to 54 days. Ventilation rates for at least two periods were obtained for six of the eight tanks, and ventilation rates for seven different (consecutive) time periods were determined for one of the tanks, Tank U-103. Table S.1 contains the best estimates of average headspace ventilation rates determined for the eight tanks studied.

Table S.1. Summary of Tank Ventilation Rates

Tank	Tracer Gas	Time Period	Average Ventilation Rate	
			(m ³ /h)	(ft ³ /min)
A-101	He	July 9–July 15, 1997	17	10
AX-102	He	August 28–September 8, 1997	28	16
AX-103	He	February 28–March 3, 1997	42	25
BY-105	SF ₆	April 17–May 8, 1997	27	16
C-107	He	February 21–March 21, 1997	1.9	1.1
S-102	SF ₆	September 24, 1996–February 11, 1997	3.8	2.2
U-103	SF ₆	February 27–July 22, 1997	3.0	1.8
	He	July 15–August 13, 1997	2.7	1.6
U-105	He	July 18–August 15, 1997	8.5	5.0

As shown in Table S.1, ventilation rates were significantly different for different tanks. Measured ventilation rates for individual periods ranged from a low of 1.7 m³/h over a 22-day period in Tank C-107, to a high of 42 m³/h over a 6-day period in Tank AX-103. Variations in the ventilation rate of a given tank for different time periods were also significant, with individual values ranging from 67% to 179% of the average value.

Acknowledgments

The authors gratefully acknowledge the support of other project staff at PNNL who contributed to the successful completion of this project. Thanks go to Jeff Edwards for cleaning and shipping and receiving of SUMMA^(a) canisters, Milt Goheen and his laboratory staff for helium analyses by mass spectroscopy, and Janet Julya for other analyses.

(a) SUMMA is a trademark of Molectrics Corporation.



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1.0 Introduction

1.1 Purpose

The purpose of this report is to detail the results of tank headspace ventilation studies using helium (He) and sulfur hexafluoride (SF₆) as tracer gases. The study was designed to meet three main objectives: 1) to determine whether tracer gases are suitable for measurement of tank ventilation rates; 2) to determine which tracer gas would be optimal for use in Hanford tanks; and 3) to obtain actual ventilation rate measurements for several tanks for different time periods within the study.

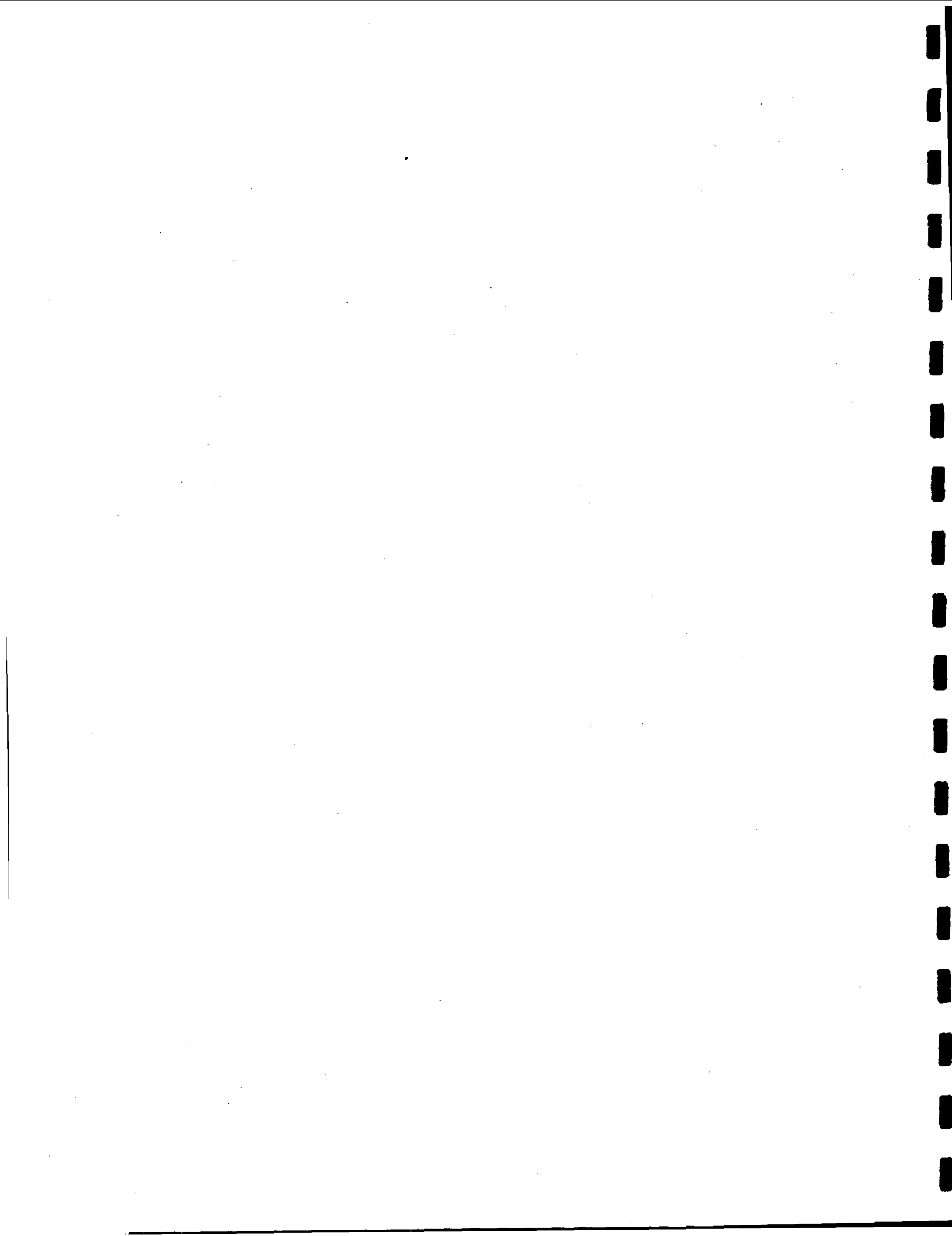
1.2 Background

Most single-shell, high-level radioactive waste tanks at the Hanford Site in Richland, Washington, are passively ventilated with the atmosphere to prevent pressurization of the tanks and accumulation of gases released by the waste. Passive ventilation rates are needed for the resolution of several key safety issues. These safety issues are associated with the rates of flammable gas production and ventilation, the rates at which organic salt-nitrate salt mixtures dry out, and the estimation of organic solvent waste surface areas. Overestimating the ventilation rate could lead to the conclusion that flammable gases are vented more quickly than they are. Underestimating the ventilation rate could result in underestimating gas production and waste dry-out rates, and potentially concluding that a tank does not have a significant amount of organic liquid waste, when indeed it does.

Direct measurement of passive ventilation rates using mass flow meters is not feasible because ventilation occurs via multiple pathways to the atmosphere (i.e., via the filtered breather riser and unsealed tank risers and pits), as well as via underground connections to other tanks, junction boxes, and inactive ventilation systems. Ventilation rates for a small number of flammable gas generating tanks have been indirectly measured by monitoring the decrease in hydrogen concentration after a significant amount of trapped hydrogen was released by the waste (Wilkins et al. 1997). Although this method provides credible results, it cannot be applied to tanks that do not trap and release significant amounts of hydrogen or tanks without hydrogen monitoring instrumentation. Numerical models have been developed and used to estimate passive ventilation rates^(a); however, such models depend heavily on estimates of physical parameters (e.g., an effective cross-section area of the ventilation pathways) that cannot be specified a priori. Simple models have also been suggested to bound the ventilation rate (Epstein et al. 1994; Cowley et al. 1997), but these do not agree with rates derived from hydrogen monitoring data (Huckaby and Sklarew 1997).

The tracer gas method discussed in this report provides a direct measurement of the rate at which gases are removed by ventilation, and an indirect measurement of the ventilation rate. The tracer gas behaves as a surrogate of the waste-generated gases, except that it is only diminished via ventilation, whereas the waste gases are continuously released by the waste and may be subject to depletion mechanisms other than ventilation.

(a) Personal communications with Don Ogden, Numatec Hanford Corp., and Zen Antoniak, PNNL.



2.0 Method Description

2.1 Overview

The tracer gas method for measurement of tank ventilation rates requires the injection of a small quantity of a tracer gas into the tank headspace, and that samples from the headspace be collected periodically and analyzed to determine the tracer gas concentration as a function of time. Provided key assumptions are satisfied, the tracer gas concentration will decrease logarithmically with time, and the ventilation rate of the tank can be calculated from the headspace volume and the rate at which the tracer gas concentration decreases.

A key assumption in the method is that the only significant change in tracer gas concentration is loss via ventilation. This implies that the tracer gas be chemically and radiolytically inert and insoluble in the waste. In this study two different tracer gases were used in the initial five tanks to verify, by comparison, their behavior in the tank headspaces.

A second key assumption in the method is that the tracer gases are essentially uniformly distributed within the tank headspace. First, this requires that the injected tracer gas be well mixed within the headspace before its baseline concentration is measured. And second, this requires that fresh air brought into the headspace via the ventilation system be mixed within the headspace in a relatively short time period (i.e., mixing should occur within several hours). The need for this assumption is twofold; first, the mathematical description of tracer gas concentration with time is greatly simplified, and second, a uniform distribution of tracer gas throughout the headspace allows representative samples to be collected via any available tank riser. The validity of this assumption was examined using a numerical model before the tracer gas method was tested on a waste tank,^(a) and tests in Tank S-102 to demonstrate mixing was occurring (see Section 5.1).

2.2 Selection of Tracer Gases

Both SF₆ and He were used as tracer gases in this study. These gases are commonly used for tracer tests because of their chemical stability, lack of toxicity, and low solubility in water.^(b) Both are very difficult to ionize and He is not subject to radiolysis. Both are also easily sampled and measured.

An advantage of SF₆ is that it is purely anthropogenic, so that the natural background levels are essentially zero (Olschewski et al. 1995). Analytical detection limits are also very low using electron capture detection instrumentation. However, because the effect on SF₆ of the high radiation levels and high alkalinity present in Hanford tanks was not known, it was necessary to first verify the stability of SF₆ under tank conditions.

(a) Modeling results were transmitted via a letter report: Antoniak, ZI and KP Rectnagle. 1996. *Modeling Tracer Gas Concentrations in Single Shell Tank 241-S-102 Dome*, TWSFG96.23, Pacific Northwest National Laboratory, Richland, Washington.

(b) Cosgrove and Walkley (1981) report the solubility of SF₆ in water at 25°C to be 4.4 x 10⁻⁶ mole fraction in water. Hildebrand and Scott (1950) report the solubility of He in water at 25°C to be 7.0 x 10⁻⁶ mole fraction.

2.3 Selection of Tanks

Waste tanks were selected for study on the bases of their associations with specific safety issues (e.g., potential waste dry-out and flammable gas generation), and for their potential to have high passive ventilation rates. The initial tank studied, Tank S-102, was selected for a demonstration of the tracer gas method of measuring ventilation rates, a test of the rate at which the tracer gases were mixed, and a comparison of SF₆ and He as tracers in the waste tanks.

Although the comparison of SF₆ and He in Tank S-102 was incomplete,^(a) results from Tank S-102 were deemed adequate to extend the study to four additional tanks: Tanks C-107, AX-103, U-103, and BY-105. These tanks were primarily selected as tanks that might have high ventilation rates, in an effort to establish an upper bounding value for passive ventilation rates. Both SF₆ and He were injected into these tanks to complete the comparison of the two tracer gases. The tracer gas method was also applied to Tanks A-101, U-105, and AX-102. These tanks were selected primarily to address specific safety issues.

(a) The schedule for collecting tracer gas samples was affected by administrative controls, and an insufficient number of samples containing appreciable He concentrations were obtained. Details and specific recommendations are given in the letter report, J Huckaby, 1997, *Preliminary Letter Report of Tracer Gas Testing Conducted to Determine Tank 241-S-102 Ventilation Rates and Demonstrate Headspace Mixing*, TWSFG97.34, Pacific Northwest National Laboratory, Richland, Washington.

3.0 Sample Collection and Analysis

3.1 Tracer Gas Injection and Sampling

Tracer gases were released into the tank headspaces via tubing that extended through a tank riser to a location approximately halfway between the waste surface and the bottom of the riser. Flow-through or purgeable gas cylinders were prepared by the laboratory with known amounts of SF₆ for injection into each tank. Typically a volume of 200 mL of SF₆ (at atmospheric pressure) was sufficient to reach the desired initial headspace concentration of about 150 parts per billion by volume (ppbv). Because larger quantities of He were needed to reach the desired initial concentration, He was supplied directly from a commercial high-pressure gas cylinder. He volumes of about 250 to 500 L (at atmospheric pressure) were injected into Tanks S-102, C-107, AX-103, and U-103 to attain desired initial concentrations of 100 to 400 parts per million by volume (ppmv). Other tanks received larger quantities of He (as much as 7,300 L) to increase the duration of the study. The He tracer gas was injected through the SF₆ gas cylinder (to purge all the SF₆ from the cylinder) and into the tank headspace via the same tubing as used for injection of SF₆. An in-line orifice was used to maintain flow rates below safety guidelines. After introduction of the tracer gas, the tank headspaces were allowed to stabilize for 1 day before baseline (time zero) samples were collected.

Headspace samples were collected in evacuated SUMMA™ passivated stainless steel canisters from the mid-elevation of the headspace via stainless steel or PFA Teflon tubing. The tubing was purged with headspace air using a combustible gas meter at a nominal flow rate of 500 mL/min for at least 5 minutes prior to collecting samples. Samples were collected in duplicate to provide an indication of sampling precision and a spare sample in case of problems.

Two headspace samples were also collected prior to injection of the tracer gas to establish background levels of SF₆ and He in each tank headspace. These background samples contained He levels of 4 to 5 ppmv and SF₆ levels below the analytical quantitation limit of 0.032 ppbv. These concentrations are essentially the same as present in ambient air, and evidently neither He nor SF₆ are produced in significant quantities by the tank waste.

Typically samples were collected at 1 day and 7 days after tracer gas injection to establish an approximate ventilation rate for each tank. If this initial ventilation rate was high, the sampling schedule was accelerated to complete sampling before the tracer gas concentrations dropped below measureable levels. If the initial ventilation rate was low or moderate, the second set of samples were collected approximately 30 days after tracer gas injection. Sampling dates and times are listed in the appendix for the eight tanks studied.

Samples were also collected, in duplicate, 15, 30, and 60 minutes after injection of SF₆ and He into Tank S-102 to determine whether rapid mixing of the tracer occurred in the headspace. These samples were collected through a different riser than the riser used to introduce the tracers, a distance of about 4 m away from the injection point.

Table 3.1 lists the eight tanks studied, their estimated headspace volumes, and the approximate volumes (at local atmospheric pressure) of He and SF₆ tracer gases injected. Headspace volumes listed are those of Palmer et al. (1996).

Table 3.1. Headspace and Tracer Gas Volumes

Tank	Estimated Headspace Volume (m ³)	Tracer Gas Injection Date	Injected Volume of He (m ³)	Injected Volume of SF ₆ (mL)
A-101	1,220	July 8, 1997	2.8	n.a.
AX-102	4,637	August 27, 1997	6.2	n.a.
AX-103	4,357	February 24, 1997	0.50	870
BY-105	2,323	April 16, 1997	0.31	620
C-107	2,342	February 20, 1997	0.34	470
S-102	1,921	September 23, 1996	0.25	160
U-103	1,451	February 26, 1997	0.23	290
		July 14, 1997	6.7	n.a.
U-105	1,625	July 17, 1997	7.3	n.a.

n.a. = not applicable because SF₆ was not injected into the tank headspace.

3.2 Sample Analysis

3.2.1 SF₆ Analysis

SF₆ was analyzed by a gas chromatograph (GC) with an electron capture detector (ECD) according to PNNL Technical Procedure SF6-97. SUMMA™ canister samples were first pressurized with ultra-high purity nitrogen to exactly double the initial filling pressure. A 2 mL gas sample loop was filled with a gas sample taken by syringe from the SUMMA™ canister and injected onto a 2.4 m molecular sieve (HP5Å 45/60 mesh) GC column. A 15 m x 0.53 mm ID HP-5 (cross linked 5% phenyl silicone) column was used to generate back pressure on the split-splitless injector. The argon/5% methane carrier gas flow rate was 22 mL/min. The oven temperature was held isothermal at 40°C for the 8-minute analysis. Column back pressure was held at 3.6 psi for the first 4 minutes and increased to 15 psi for the last 4 minutes of the run time to facilitate purging of the column. The ECD was held at 250°C.

Commercially purchased calibration standards with concentrations of SF₆ certified to 5% accuracy were used to calibrate the GC-ECD. Three calibration ranges were needed to minimize the effects of the non-linear response of the ECD to this strongly electron-capturing compound (Farwell et al. 1981). The three calibration curves ranged from 0.030 to 1 ppbv (linear regression based on 3 points), 1 to 20 ppbv (quadratic fit based on 4 points), and 20 to 160 ppbv (cubic fit based on 5 points).

The GC-ECD system used for SF₆ had a relative standard deviation (RSD) of less than 8% for each of the three calibration ranges. The estimated instrument detection limit was 0.006 ppbv and the estimated quantitation limit was 0.032 ppbv. The SF₆ concentrations measured in the tanks and listed in

the appendix were estimated to be accurate to within 10% of their true values based on method performance tests.

3.2.2 He Analysis by MS

Selected samples were analyzed for He by a high sensitivity mass spectrometer (MS), equipped with both a Faraday cup detector (operated in the current mode and linear to 1 part in 100,000) and a secondary electron multiplier (SEM) which was used for ion counting. In this method, which was based on PNNL Procedure ALO 284 Rev. 1 and applicable to all ideal gases, all gas species were scanned, their relative amounts determined, and the results normalized.

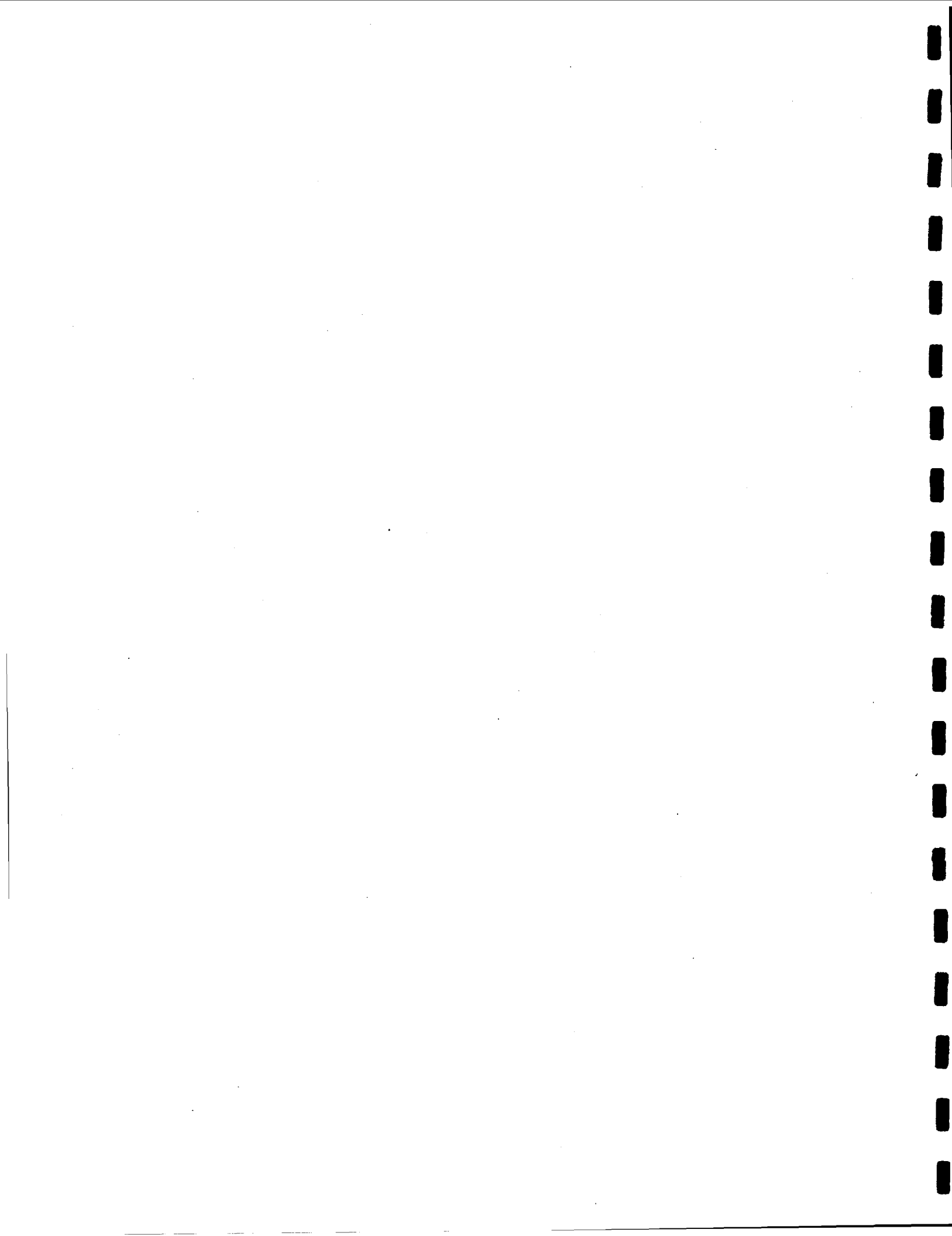
The MS system used for He has a replicate precision of about 2% RSD. The He concentrations measured in the tanks and listed in Table A.1 of the appendix are estimated to be accurate to within ± 5 ppmv of their true values. The instrument detection limit is typically less than 1 ppmv.

3.2.3 He Analysis by GC-TCD

Helium was analyzed by a micro GC with a thermal conductivity detector (TCD) (P200H Micro GC from MTI Analytical Instruments, Inc.) as described in PNNL Technical Procedure HE-97. SUMMA™ canister samples were first pressurized with ultra-high purity nitrogen to exactly double the initial filling pressure. Gas samples were taken by syringe from the SUMMA™ canister and injected (50 millisecond injection time) onto a 4 m x 0.32 mm ID 5Å molecular sieve column. High purity argon was used as the carrier gas; column pressure was 15 psig. The oven was held isothermal at 35°C for the 1-minute analysis time.

A commercially prepared and certified mixture of 2,000 ppmv He in nitrogen was diluted using an Entech 4569 Dynamic Dilution System (or equivalent) to prepare a series of calibration standards to concentrations as low as 2 ppmv. The GC-TCD system used for He was highly linear from 2 ppmv to 2,000 ppmv and had a replicate precision of about 1% RSD or slightly higher (less than 5%) near the quantitation limit. The instrument detection limit was 1.0 ppmv and the estimated quantitation limit was 2 ppmv.

The GC-TCD method was used to measure He concentrations in samples from Tanks A-101, U-103, and U-105. Results are listed in the appendix.



4.0 Calculation of Ventilation Rates

4.1 Ventilation Rate Calculations

Given a well-mixed headspace, the decrease in tracer concentration with time is proportional to its concentration:

$$\frac{dC}{dt} = -\frac{Q}{V}C \quad (1)$$

where C is the concentration of the tracer, Q is the volumetric ventilation rate, V is the headspace volume, and t is time. This equation can be solved for the ventilation rate, Q , between any two sample events:

$$Q = \frac{V}{(t_i - t_j)} \ln\left(\frac{C_j}{C_i}\right) \quad (2)$$

where C_i and C_j are the concentrations of the tracer gas at any two different times t_i and t_j , respectively. When concentration is plotted against time on a log-linear scale, the slope of the resulting line is proportional to the estimated ventilation rate. Nonlinearity of more than two points may be caused by sampling and analytical measurement errors or by changes in the ventilation rate.

Averaged tracer gas concentrations for each sampling event and ventilation rates between each consecutive pair of sampling events are listed in Table 4.1 for Tanks A-101, AX-103, BY-105, C-107, S-102, U-103, and U-105. Ventilation rates listed were calculated with equation (2) and headspace volumes given in Table 3.1. The average ventilation rate is based on the initial and final tracer measurements.

4.2 Statistical Analysis

Equation (2) can be applied between any two points in time at which tracer gas concentrations are known. To apply it to more than two points and account for residual errors, it may be rewritten

$$\ln(C_i) = -\frac{Q}{V}t_i + \ln(C_0) + \epsilon_i \quad (3)$$

where C_0 is the tracer gas concentration at time $t = 0$, and ϵ_i is the residual error associated with the sampling and analytical variability of C_i . A plot of equation (3) yields a line with slope of $-Q/V$ and an intercept of $\ln(C_0)$.

Table 4.1. Summary of Ventilation Rates by Tank and Sample Date

Tank	Tracer Gas	Sample Date	Concentration (He ppmv, SF ₆ ppbv)	Ventilation Rate (m ³ /h)	95% LCL (m ³ /h)	95% UCL (m ³ /h)
A-101	He	09-Jul-97	1,155	-	-	-
		15-Jul-97	153	17	12	23
AX-102	He	08-Aug-97	712	-	-	-
		02-Sep-97	297	34	30	39
		08-Sep-97	148	23	20	25
		Average		28	25	30
AX-103	He	25-Feb-97	77	-	-	-
		03-Mar-97	20	42	33	50
	SF ₆	25-Feb-97	119	-	-	-
		03-Mar-97	4.6	101	94	107
BY-105	He	17-Apr-97	102	-	-	-
		23-Apr-97	10.5	36	31	40
		Average		26	23	29
	SF ₆	17-Apr-97	204	-	-	-
		23-Apr-97	17.4	38	36	41
		08-May-97	0.56	23	21	24
C-107	He	21-Feb-97	180	-	-	-
		27-Feb-97	155	2.4	0.0	8.2
		21-Mar-97	105	1.7	1.6	1.9
		Average		1.9	0.6	3.2
	SF ₆	21-Feb-97	179	-	-	-
		27-Feb-97	77.7	14	12	15
		21-Mar-97	2.77	15	14	16
Average		15	14	16		
S-102	He	24-Sep-96	122	-	-	-
		30-Sep-96	80	5.7	2.7	8.8
		11-Oct-96	59	2.2	0.2	4.1
		Average		3.3	2.1	4.6

Table 4.1. (contd)

Tank	Tracer Gas	Sample Date	Concentration (He ppmv, SF ₆ ppbv)	Ventilation Rate (m ³ /h)	95% Lower Confidence Level (m ³ /h)	95% Upper Confidence Level (m ³ /h)
S-102	SF ₆	24-Sep-96	102	-	-	-
		30-Sep-96	79.6	3.3	2.8	3.8
		11-Oct-96	51.6	3.1	2.4	3.8
		19-Dec-96	1.5	4.1	3.8	4.5
		11-Feb-97	0.14	3.5	3.2	3.7
		Average		3.8	3.6	4.1
	He	27-Feb-97	158	-	-	-
		06-Mar-97	98	4.1	2.7	5.5
		31-Mar-97	19	4.0	2.5	5.5
		09-Apr-97	8.0	5.6	1.6	9.7
		Average		4.3	3.9	4.7
U-103 (1)	SF ₆	27-Feb-97	193	-	-	-
		06-Mar-97	108	4.9	4.4	5.5
		31-Mar-97	20.1	4.0	3.8	4.3
		09-Apr-97	9.40	5.2	4.5	5.8
		28-May-97	1.12	2.6	2.4	2.8
		14-Jul-97	0.19	2.2	2.1	2.4
		22-Jul-97	0.13	3.0	2.1	3.9
		Average		2.9	2.7	3.1
U-103 (2)	He	15-Jul-97	3,473	-	-	-
		22-Jul-97	2,315	3.5	3.3	3.7
		13-Aug-97	962	2.4	2.2	2.6
		Average		2.6	2.4	2.9
U-105	He	18-Jul-97	2,800	-	-	-
		24-Jul-97	1,451	7.4	6.9	8.0
		15-Aug-97	84	8.8	8.2	9.3
		Average		8.6	8.1	9.1

Average = breathing rate from time 0 to final sampling event.

LCL = lower confidence limit.

UCL = upper confidence limit.

The parameters in the above linear equation can be estimated using simple linear regression (least squares algorithm) described in Draper and Smith (1981). Denoting

$$\beta_0 = \ln(C_0)$$

$$\beta_1 = -\frac{Q}{V}$$

equation (3) may be rewritten

$$\ln(C_i) = \beta_1 t_i + \beta_0 + \epsilon_i \quad (4)$$

The parameters of primary interest in this model are β_1 and its variance, $\text{var}(\beta_1)$. The ϵ_i are also of interest because they are used in the calculation of the $\text{var}(\beta_1)$ estimate. The regression reference derives and presents the formulas for these parameter estimates.

Tank headspace volumes, given in Table 3.1, were assumed to have a $\pm 5\%$ uncertainty. Assuming that the estimated headspace volumes are normally distributed with a mean at the true value, and that the $\pm 5\%$ uncertainty defines the width of the 95% confidence interval, the estimated variance of V is given by

$$\text{var}(\hat{V}) = 0.025 \hat{V}$$

where hats (^) have been used to indicate the quantity is an estimate. The estimated ventilation rate and associated estimated variance are given by

$$\hat{Q} = -\beta_1 \hat{V}$$

$$\text{var}(\hat{Q}) = \hat{V}^2 \text{var}(\beta_1) + \beta_1^2 \text{var}(\hat{V}) + \text{var}(\hat{V}) \text{var}(\beta_1)$$

The use of this variance formula assumes that headspace volume, V , and the slope, β_1 , are independent. These estimators are discussed and presented in Mood et al. (1974).

Finally, the formula for the 95% confidence interval for the vent rate estimate is

$$\hat{Q} \pm t_{0.975, df} \text{var}(\hat{Q})$$

where t is the 97.5 percentile from a t-distribution with df degrees of freedom. The degrees of freedom is equal to the number of analytical results in the regression fit, less one for the slope and one for the intercept.

5.0 Discussion of Results

5.1 Headspace Mixing Test

The assumption that gases are mixed quickly in the waste tank headspaces was tested in the first tank studied, Tank S-102. A numerical model was used prior to the release of tracer gases in Tank S-102 to predict approximate mixing rates. When the waste surface was assumed to be 1.1 °C warmer than the concrete dome (a condition that promotes mixing by thermally driven convection), the three-dimensional model indicated the tracers would be fully dispersed through out the headspace within about 30 minutes after the tracer gas release.^(a)

Model predictions were tested by collecting three pairs of samples from a point about 4 m away from the injection point,^(b) approximately 15, 30, and 60 minutes after the He and SF₆ tracer gases were injected. Table 5.1 lists the collection times and analytical results for these three sets of samples along with the same data for samples collected the following day. Significant levels of both tracers were found in the samples within 15 minutes after the injection was complete, and both tracers were essentially at their final concentrations at the sampling point within one hour after injection. Tracer concentrations measured 1-day after injection agree well with that calculated from the estimated headspace volume and quantities of tracer released.

Given this evidence for vigorous mixing, inhomogeneities produced by the influx of fresh air during normal ventilation would be expected to be restricted to a small region near the influx. Considering that the highest observed ventilation rates result in less than a 10% turnover of the headspace volume each day, the rate at which fresh air would be mixed within the headspace is very rapid, and the distribution of tracer gases can be assumed to be essentially uniform at any given time.

5.2 Comparison of He and SF₆ Tracer Gases

Comparison tests of He and SF₆ were conducted in five of the eight tanks to determine the stability of SF₆ under the high radiation/high alkaline conditions present in the Hanford waste tanks. Table 5.2 summarizes the directly comparable He and SF₆ ventilation rate data.

As indicated in Table 5.2, in Tanks BY-105, S-102, and U-103, the He and SF₆ concentrations decrease at the same rate and the ventilation rates based on each tracer agree well. Figure 5.1 plots the logarithm of tracer concentrations as a function of time for Tank U-103. The disconnect in He results occurs because He was injected into this tank on two separate dates. Note that the slopes of the lines drawn between the SF₆ data points and between the He data points are very similar, indicating their

(a) Letter report: Antoniuk, ZI and KP Rectnagle. 1996. *Modeling Tracer Gas Concentrations in Single Shell Tank 241-S-102 Dome*, TWSFG96.23, Pacific Northwest National Laboratory, Richland, Washington.

(b) The tracer gases were injected via a stainless steel probe mounted in riser 7, and samples were collected via a similar probe mounted in riser 1.

Table 5.1. Results of Tracer Gas Mixing Test

Sample Identifier	Date and Time	Elapsed Time (h:min)	He (ppmv)	SF ₆ (ppbv)
S6094-A01.085	9/23/96 11:00	NA	5	< 0.01
S6094-A02.246	9/23/96 11:03	NA	4	< 0.01
Injection Begun	9/23/96 11:09	NA	NA	NA
Injection Stopped	9/23/96 11:25	0:00	NA	NA
S6094-A03.298	9/23/96 11:40	0:15	50	108
S6094-A04.299	9/23/96 11:42	0:17	50	109
S6094-A05.334	9/23/96 11:55	0:30	160	105
S6094-A06.335	9/23/96 11:56	0:31	200	113
S6094-A07.336	9/23/96 12:25	1:00	120	96
S6094-A08.337	9/23/96 12:26	1:01	120	108
S6094-B02.338	9/24/96 10:48	23:23	120	103
S6094-A25.105	9/24/96 11:37	24:12	125	101
S6107-A26.106	9/24/96 11:41	24:16	137	102

NA = not applicable

In contrast, in Tanks AX-103 and C-107, SF₆ decreases at a much faster rate than He, and the ventilation rate calculated from SF₆ data is greater than two times that calculated from He data. Figure 5.2 plots the logarithm of tracer concentrations as a function of time for Tank C-107. The slopes of lines drawn through the He and SF₆ data are clearly different, and illustrate that SF₆ is being lost at a much higher rate than He. Because He and SF₆ were injected and sampled on the same dates (He and SF₆ analyses were performed on the same samples), the discrepancy in ventilation rates cannot be attributed to a temporal difference. Helium was present in the tank prior to injection at normal background levels; therefore, the higher concentrations of He cannot be attributed to an abnormally high background.

It appears that contrary to expected behavior, the SF₆ must be less inert or more soluble in the waste than anticipated. In the literature, the usefulness of SF₆ as a tracer has been based on the fact that it is inert and relatively insoluble in water. However, the Hanford tanks provide a unique situation for the use of tracers. The presence of high levels of radioactivity in the tank may, in fact, affect the stability of the SF₆. Because of the presence of fission product-derived beta emitters, including ⁹⁰Sr, ¹³⁷Cs, ⁹⁹Tc, and ³H, these tanks should have a substantial supply of thermal electrons present. In addition, radiolysis of water produces hydrated electrons (Asmus et al. 1970) that would be available in the tank to react with SF₆.

Table 5.2. Comparison of He and SF₆ Ventilation Rates

Tank	Period	He Ventilation Rate (m ³ /h)	SF ₆ Ventilation Rate (m ³ /h)
AX-103	Day 1–Day 5	41.7	100.5
BY-105	Day 1–Day 7	37.9	38.4
C-107	Day 1–Day 7	2.4	14.0
	Day 7–Day 29	1.7	15.0
	Day 1–Day 29	1.9	15.0
S-102	Day 1–Day 7	5.7	3.3
	Day 7–Day 17	2.2	3.1
	Day 1–Day 17	3.4	3.2
U-103	Day 1–Day 7	4.1	4.9
	Day 7–Day 32	4.0	4.0
	Day 32–Day 41	5.6	5.2
	Day 1–Day 41	4.4	--

A literature review indicated that SF₆ is frequently used in radiation studies as an electron scavenger (Niedzielski and Gawłowski 1973a). For example, in radiolysis of hydrocarbons in the presence of SF₆, the SF₆ acts as an electron scavenger and compounds of the general formula RSF₅ are formed (Gawłowski and Herman 1974). Foster and Beauchamp (1975) note that “contrary to the usual assumption that it is unreactive, SF₆ formed by electron attachment” in radiolytic systems “is found to undergo rapid bimolecular reactions with molecules possessing acidic hydrogen” (including formic and acetic acids); this paper does not consider what may happen in an alkaline environment. However, Asmus et al. (1970) have shown that alkaline hydrolysis of SF₄ (formed from radiolytic decomposition of SF₆) has a high rate constant ($7 \times 10^9 \text{ M}^{-1}\text{sec}^{-1}$), higher even than the rate constant in acidic solution. The yield of radiolytic decomposition of SF₆ has been found to depend linearly upon dose (Niedzielski and Gawłowski 1973b).

Though no definitive explanation for the accelerated loss of SF₆ was determined, there is an apparent correlation between the loss of SF₆ and tank headspace temperatures. Of the tanks studied, Tanks AX-103 and C-107 (in which He and SF₆ results did not agree) are significantly hotter than the other tanks. The higher temperatures of Tanks AX-103 and C-107 may be associated with higher beta fields in their headspaces. Higher beta fields may in turn cause unexpectedly high rates of SF₆ radiolysis, because SF₆ has a very large electron (beta particle) capture cross section. The apparent correlation of SF₆ loss with tank temperature is inconsistent with the premise that SF₆ is being absorbed by the aqueous waste, because the solubility of SF₆ in the waste is expected to decrease with increasing temperature.

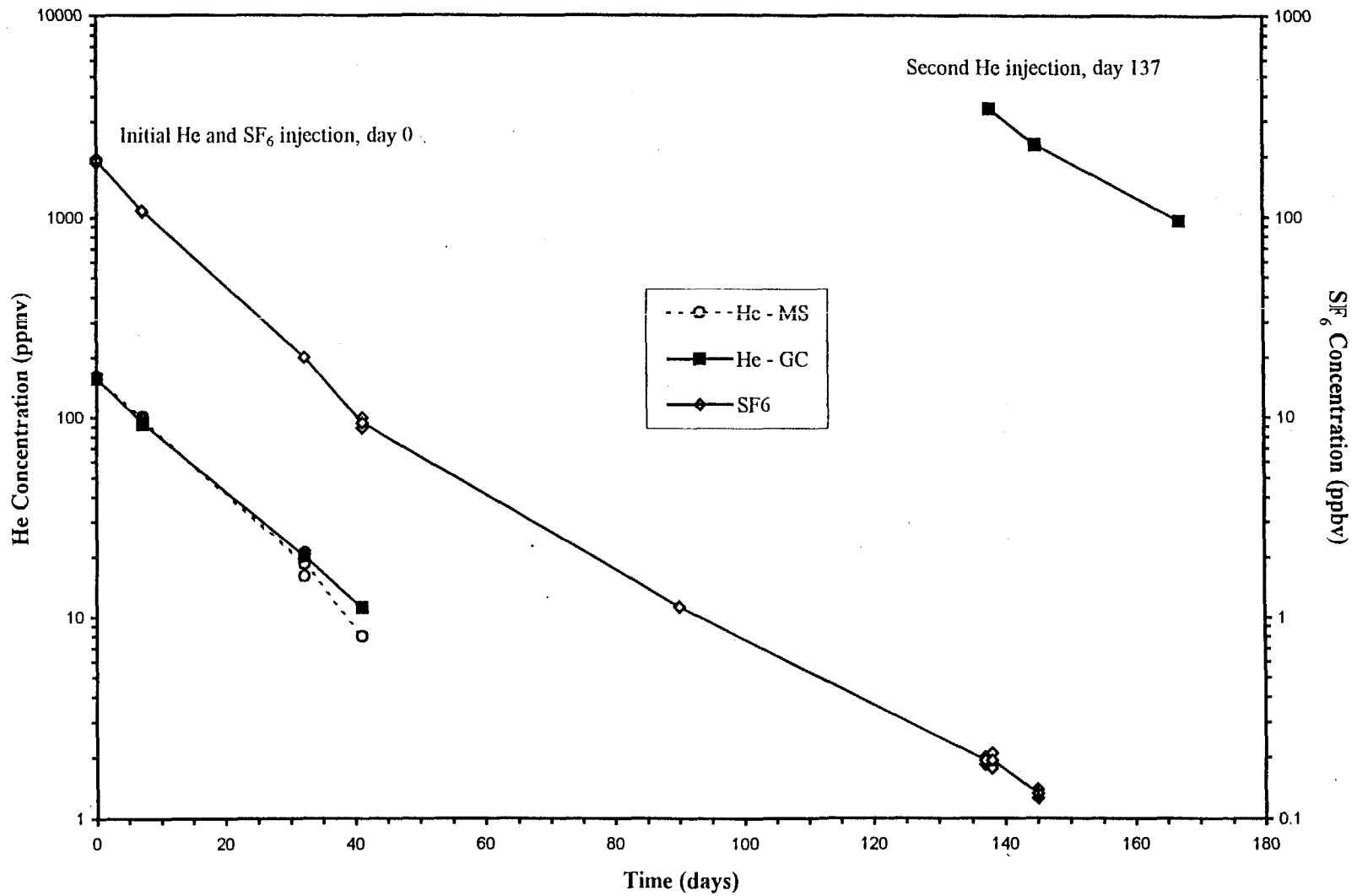


Figure 5.1. Tank U-103 He and SF₆ Concentrations

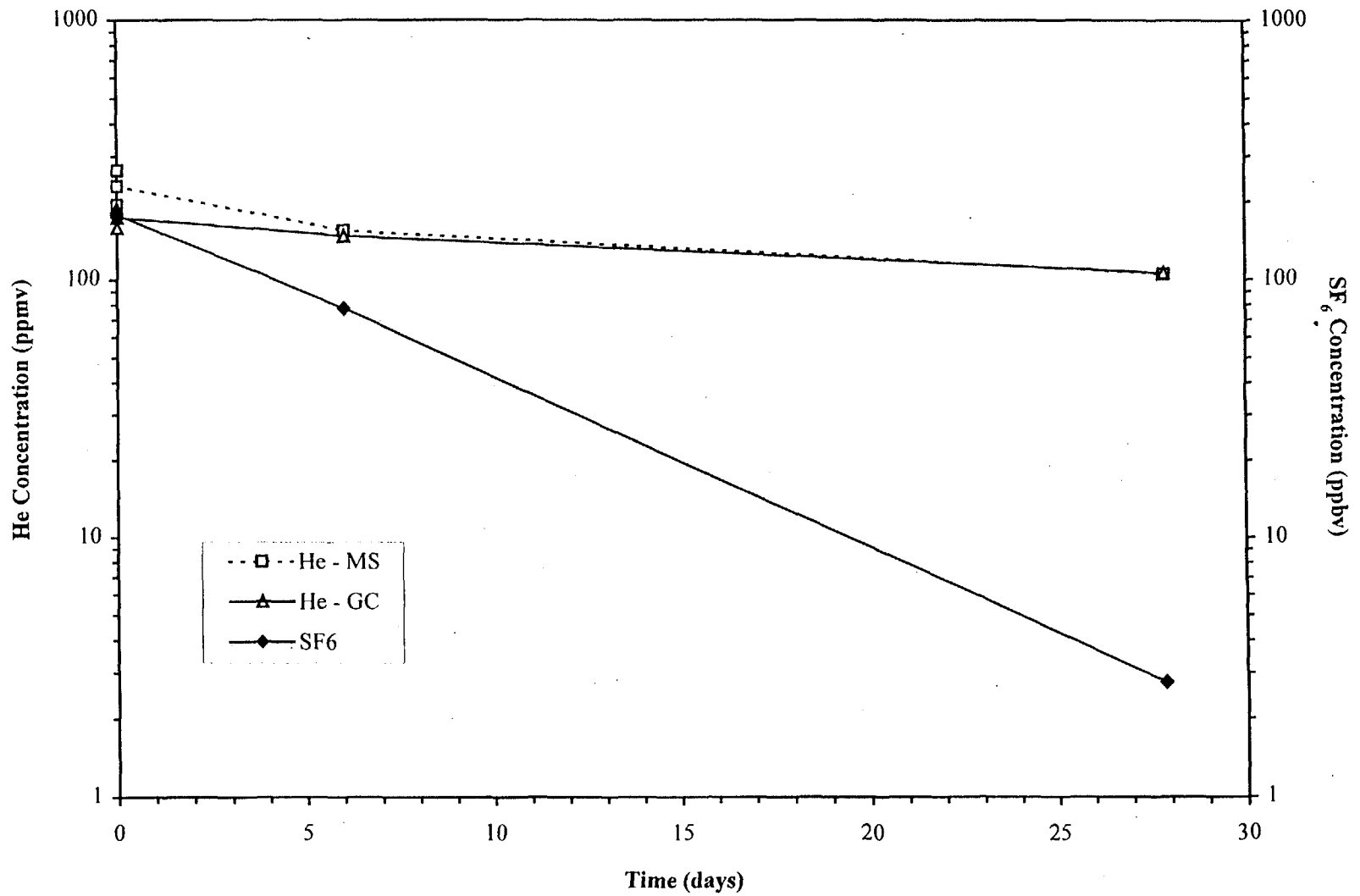


Figure 5.2. Tank C-107 He and SF₆ Concentrations

Because of the discrepancy between He and SF₆ results, SF₆ was not used as a tracer gas in the most recently studied tanks, Tanks A-101 and U-105, nor in the second injection of tracer gas in Tank U-103. Ventilation rates based on SF₆ data (e.g., the majority of data for Tank S-102) represent upper bounding estimates, because if SF₆ had also been depleted by some other mechanism, an erroneously high ventilation rate would be calculated. Based on the initial agreement of He and SF₆ ventilation rates in Tanks S-102, BY-105, and U-103, it appears likely that the two tracer gases would give essentially equivalent results in these tanks.

5.3 Tank AX-102 Sample Contamination Problems

Anomalous results were obtained for the initial Tank AX-102 tracer study. This tank received an injection of He gas on August 4, 1997, and the standard background, 1-day, and 7-day sample sets were collected. Unexpectedly high He concentrations of about 50,000 ppmv (compared with an expected maximum of about 2,500 ppmv) were found in each of the initial three sets of Tank AX-102 samples. An investigation of the anomalous results indicated that the SUMMA™ canisters must have been contaminated with He during the canister cleaning operation (before being sent to the field). The contamination was associated with the use of He to perform a manifold leak check and a mechanical valve failure.

To prevent future contamination of the SUMMA™ canisters with He, two procedural changes were implemented: 1) the use of He in the cleaning process was eliminated and 2) the vacuum bellows valves on all SUMMA™ canisters are to be tested and any suspect valves are to be replaced. Helium was injected into Tank AX-102 again in late August, and the background, 1- and 5-day samples were found to contain expected concentrations of He.

5.4 Ventilation Rate Variability

As shown in Table 4.1, ventilation rates were significantly different for different tanks. Measured ventilation rates for individual periods ranged from a low of 1.7 m³/h over a 22-day period in Tank C-107, to a high of 42 m³/h over a 6-day period in Tank AX-103. Variations in the ventilation rate of a given tank for different time periods was also significant, with individual values ranging from 67% to 179% of the average value.

Headspace ventilation rates of Tank S-102 were measured over a 140-day period beginning in early fall and ending after the coldest part of the winter had passed. As shown in Table 4.1, the four calculated ventilation rates based on SF₆ concentrations in Tank S-102 varied only from 3.1 to 4.1 m³/h. However, based on the narrow ranges of 95% upper and lower confidence limits given in Table 4.1, these ventilation rate differences between the different periods appear to be significant. The highest average ventilation rate observed in Tank S-102 occurred between mid-October and mid-December. Passive ventilation via air leaks in riser flanges and pit covers may also have been affected by snow cover during the mid-December to mid-February period.

Tank U-103 was also studied over an extended time period, from late February to mid-August, and it is currently expected that the study will be continued through October. Both He and SF₆ were injected

into Tank U-103 in late February and more He was injected in mid-July to extend the study. This was undertaken to examine seasonal changes in passive ventilation rates. The planned study is incomplete, but the available data suggest an inverse correlation between ventilation rate and ambient temperatures. Table 5.3 presents a comparison of the average ambient air temperature (third column) and ventilation rate (last column) for each sequential set of tracer samples. Although fluctuations in the ventilation rate were observed, the ventilation rate has generally decreased as ambient temperatures have risen.

It is currently thought that passive ventilation is partially driven by the bouyancy difference between warm air in the headspace and cool ambient air. If that is correct, the ventilation rate is more appropriately correlated with the difference between ambient and headspace temperatures. The average headspace temperature for Tank U-103 is included in Table 5.3 (fourth column), as well as the difference between average ambient and headspace temperatures (fifth column). Comparison of the temperature differences with the ventilation rates suggests that while ventilation rate does decrease as the temperature difference decreases, it approaches and fluctuates about a minimum value, and is not decreased by further decreases in the temperature difference.

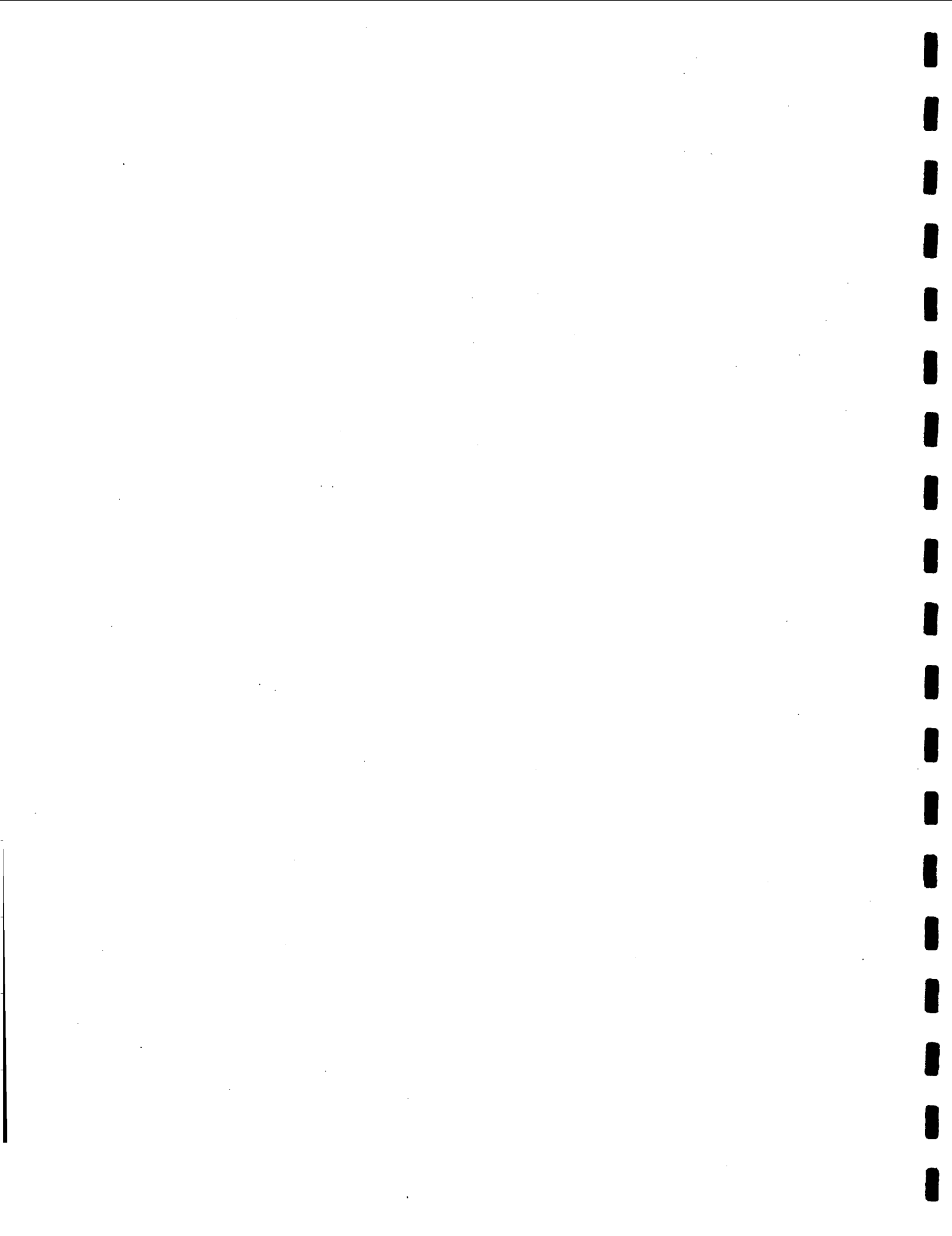
Table 5.3. Tank U-103 Ventilation Rates and Temperatures

Tracer Gas	Time Period (1997)	Average Ambient Air Temperature ^(a) (°C)	Average Headspace Temperature ^(b) (°C)	Temperature Difference (°C)	Average Ventilation Rate (m ³ /h)
SF ₆	February 27–March 6	41.5	69.9	28.4	4.9
	March 6–March 31	48.5	69.6	21.1	4.0
	March 3–April 9	47.6	69.8	22.2	5.2
	April 9–May 28	59.4	70.9	11.5	2.6
	May 28–July 14	69.4	74.5	5.1	2.2
	July 14–July 22	79.4	76.7	-2.7	3.0
He	July 14–July 22	79.4	76.7	-2.7	3.5
	July 22–August 13	79.5	78.0	-1.5	2.4

(a) Average ambient air temperatures are based on hourly readings obtained from Hanford Meteorological Station.

(b) Average headspace temperatures are based on averages of the daily readings of two thermocouples in the headspace of Tank U-103. Readings were obtained from the Tank Characterization Database.

Tracer sample collection from Tank U-103 was timed to address the impact of push-mode core sampling operations on passive ventilation rates. Samples were collected on March 31, just prior to the start of coring operations, and again on April 9, after coring operations were complete. As shown in Table 5.3, a small increase in the ventilation rate was observed during the period of coring operations, and this may have been caused by the coring operations. However, this increase is comparable to fluctuations observed in the other tanks studied (which were not subject to comparable activities), and may not have been caused by coring operations.



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Appendix

Analytical Results

Table A.1. Mean Adjusted Analytical Results

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
A-101	He - MS	7/9/97 10:23	V0001-B01.149	1,155
		7/9/97 10:30	V0001-B02.223	1,155
		7/15/97 10:48	V0001-C02.071	153
A-101	He - GC	7/9/97 10:23	V0001-B01.149	1,185
		7/9/97 10:30	V0001-B02.223	1,177
		7/15/97 10:48	V0001-C02.071	139
AX-102	He-GC	8/28/97 9:10	V0003-B01.123	760
		8/28/97 9:15	V0003-B02.127	664
		9/2/97 9:00	V0003-C01.220	302
		9/2/97 9:10	V0003-C02.225	292
		9/8/97 9:46	V0003-D01.230	153
		9/8/97 9:51	V0003-D02.275	143
AX-103	SF ₆	2/25/97 13:36	V7011-B01.232	120
		2/25/97 13:42	V7011-B02.247	119
		3/3/97 10:45	V7011-C01.158	4.6
		3/3/97 10:51	V7011-C02.160	4.6
AX-103	He - MS	2/25/97 13:36	V7011-B01.232	79
		2/25/97 13:42	V7011-B02.247	75
		3/3/97 10:45	V7011-C01.158	19
		3/3/97 10:51	V7011-C02.160	21
		3/24/97 11:02	V7011-D01.091	0
		3/24/97 11:07	V7011-D02.092	0
AX-103	He - GC	2/25/97 13:36	V7011-B01.232	78.7
		2/25/97 13:42	V7011-B02.247	78.7
		3/3/97 10:45	V7011-C01.158	28.3
		3/3/97 10:51	V7011-C02-160	27.8

Table A.1. (contd)

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
BY-105	SF ₆	4/17/97 9:34	V7010-B01.157	205
		4/17/97 9:40	V7010-B02.215	203
		4/23/97 14:35	V7010-C01.030	17.3
		4/23/97 14:40	V7010-C02.125	17.5
		5/8/97 9:41	V7010-D01.060	0.567
		5/8/97 9:47	V7010-D02.086	0.548
BY-105	He - MS	4/17/97 9:34	V7010-B01.157	95
		4/17/97 9:40	V7010-B02.215	85
		4/23/97 14:35	V7010-C01.030	7
		4/23/97 14:40	V7010-C02.125	9
BY-105	He - GC	4/17/97 9:34	V7010-B01.157	108.5
		4/17/97 9:40	V7010-B02.215	94.5
		4/23/97 14:35	V7010-C01.030	8.7
		4/23/97 14:40	V7010-C02.125	12.3
		5/8/97 21:41	V7010-D01.060	6.0
		5/8/97 21:47	V7010-D02.086	5.7
C-107	SF ₆	2/21/97 13:32	V7009-B01.024	182
		2/21/97 13:37	V7009-B02.027	175
		2/27/97 13:27	V7009-C01.021	78.1
		2/27/97 13:32	V7009-C02.028	77.3
		3/21/97 9:51	V7009-D01.042	2.76
		3/21/97 9:57	V7009-D02.045	2.78
		2/21/97 13:32	V7009-B01.024	195

Table A.1. (contd)

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
C-107	He - MS	2/21/97 13:37	V7009-B02.027	165
		2/27/97 13:27	V7009-C01.021	155
		2/27/97 13:32	V7009-C02.028	155
		3/21/97 9:51	V7009-D01.042	105
		3/21/97 9:57	V7009-D02.045	105
C-107	He - GC	2/21/97 13:32	V7009-B01-024	189
		2/21/97 13:37	V7009-B02.027	159
		2/27/97 13:27	V7009-C01.021	148
		2/27/97 13:32	V7009-C02.028	147
		3/21/97 9:51	V7009-D01.042	106
		3/21/97 9:57	V7009-D02.045	106
		9/24/96 10:48	S6094-B02.339	103
S-102	SF ₆	9/24/96 11:37	S6093-A25-105	101
		9/24/96 11:41	S6093-A26-106	102
		9/30/96 10:02	S6094-C01.340	80.2
		9/30/96 10:04	S6094-C02.341	78.9
		10/11/96 14:26	S6094-D01.236	50.6
		10/11/96 14:28	S6094-D02.239	52.6
		12/19/96 11:06	V6001-A04.272	1.45
		12/19/96 11:10	V6001-A05.276	1.53
		12/19/96 11:14	V6001-A06.312	1.45
		2/11/97 10:45	V7006-A04.142	0.144
		2/11/97 10:49	V7006-A05.167	0.144
		2/11/97 10:53	V7006-A06.169	0.137

Table A.1. (contd)

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
S-102	He - MS	9/24/96 10:48	S6094-B02.339	115
		9/24/96 11:37	S6093-A25-105	120
		9/24/96 11:41	S6093-A26-106	132
		9/30/96 10:02	S6094-C01.340	75
		9/30/96 10:04	S6094-C02.341	85
		10/11/96 14:26	S6094-D01.236	59
		10/11/96 14:28	S6094-D02.239	59
U-103	SF ₆	2/27/97 8:52	V7008-B01.015	198
		2/27/97 8:58	V7008-B02.016	188
		3/6/97 10:46	V7008-C01.003	107
		3/6/97 10:51	V7008-C02.020	109
		3/31/97 13:59	V7008-D01.037	20.1
		3/31/97 14:05	V7008-D02.040	20.1
		4/9/97 10:26	V7008-E01.093	10.0
		4/9/97 10:32	V7008-E02.112	8.87
		5/28/97 9:24	V7008-F01.019	1.13
		5/28/97 9:29	V7008-F01.047	1.12
		7/14/97 9:59	V0002-A01.227	0.201
		7/14/97 10:06	V0002-A02.241	0.184
		7/15/97 9:29	V0002-B01.269	0.210
		7/15/97 9:35	V0002-B02.315	0.177
		7/22/97 8:37	V0002-C01.085	0.140
7/22/97 8:42	V0002-C02.118	0.126		

Table A.1. (contd)

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
U-103	He - MS	2/27/97 8:52	V7008-B01.015	161
		2/27/97 8:58	V7008-B02.016	155
		3/6/97 10:46	V7008-C01.003	95
		3/6/97 10:51	V7008-C02.020	101
		3/31/97 13:59	V7008-D01.037	21
		3/31/97 14:05	V7008-D02.040	16
		4/9/97 10:26	V7008-E01.093	8
		4/9/97 10:32	V7008-E02.112	8
U-103	He - GC	2/27/97 8:52	V7008-B01.015	156
		2/27/97 8:58	V7008-B01.016	158
		3/6/97 10:46	V7008-C01.003	92
		3/6/97 10:51	V7008-C02.020	95
		3/31/97 13:59	V7008-D01.037	20.3
		3/31/97 14:05	V7008-D02.040	20.1
		4/9/97 10:26	V7008-E01.093	11.2
		4/9/97 10:32	V7008-E02.112	11.1
		5/28/97 9:24	V7008-F01.019	12.7
		5/28/97 9:29	V7008-F02.047	12.8
U-103	He - GC	7/15/97 9:29	V0002-B01.269	3,472
		7/15/97 9:35	V0002-B02.315	3,474
		7/22/97 8:37	V0002-C01.085	2,306
		7/22/97 8:37	V0002-C02.118	2,329
		8/13/97 8:57	V0002-D02.019	962

Table A.1. (contd)

Tank Name	Tracer Gas -- Method	Sample Date & Time	Sample Name	Adjusted Mean (ppbv SF ₆ or ppmv He)
U-105	He - GC	7/18/97 9:08	V0004-B01.246	2,809
		7/18/97 9:14	V0004-B02.256	2,792
		7/24/97 9:18	V0004-C01.031	1,423
		7/24/97 9:23	V0004-C02.338	1,481
		8/15/97 8:29	V0004-D01.020	85
		8/15/97 8:35	V0004-D02.023	83
He - MS Estimated error less than 5% or ±5 ppmv, whichever is higher.				
SF ₆ Estimated error less than 10%.				
He - MTI Estimated error less than 10%.				

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